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CHICAGO UNIV ILL DEPT OF CHEMISTRY CHEMISTRY AT ELECTRODE SURFACES. (U) DEC 77 S MAZUR

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FINAL REPORT

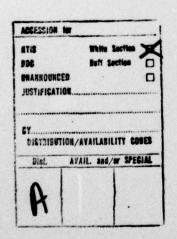
STEPHEN MAZUR

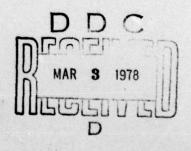
Dec. 29, 1977

U.S. ARMY RESEARCH OFFICE

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THE UNIVERSITY OF CHICAGO





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STATEMENT OF THE PROBLEM

Our objective has been to elucidate the structure of functional groups at the prismatic surfaces of graphite and related forms of carbon. We wish to make use of this functionality in order to manipulate the chemistry at the surface of carbon electrodes. Central to this project has been our proposition that prismatic surfaces of graphite, when freed of the usual surface oxides, should be composed of functionality resembling certain dehydroaromatic molecules. It would then follow that the reactivity of these surfaces should be analogous to the reactivity of the dehydroaromatics.

As a natural outgrowth of these studies, it was of importance to know how the dehydroaromatic molecules react with molecular oxygen since the reaction of oxygen with the carbon suraces is of great importance. Moreover the structure of the surface oxides remains poorly defined despite the tremendous amount of work that has been done in this field. Our approach to this problem was to prepare benzyne, a reactive dehydroaromatic, as a substituent on a cross-linked polystyrene matrix and investigate its chemistry under conditions where the possible competition of dimerization reactions would be less of a problem.

SUMMARY OF RESULTS

Most of the technical details of this work has appeared in previous progress and technical reports so only a breif outline will be given here. The surface of oxide-free carbon was shown to be quite reactive towards a variety of olefinic reagents in good agreement with the "dehydroaromatic model". By means of these reactions several new surface derivatives have been prepared and characterized, among them an electroactive functionality.

A preliminary report of these results appeared in <u>J. Am. Chem. Soc.</u>, <u>99</u> 3888, (1977). Participating personnel were Dr. Tomislav Matusinovic and Dr. Karl Cammann. Work is continuing on this project under the joint sponsorship of the U.S. Army Research Office and the National Science Foundation.

The studies of polymer-bound benzyne have yielded the following results. In keeping with our expectations the ubiquitous dimerization of benzyne is completely supressed by immobilization on the polymer matrix. Under these conditions we were able to show that the lifetime of the benzyne is substantially longer than in solution. These results were reported

I. Am. Chem. Soc., 98, 6710 (1976). In continuing work on this system we were able to identify some new reactions of the long-lived benzyne including a slow intrapolymeric reaction. Unfortunately, the reaction with oxygen has still eluded us. A full paper on this work is currently in preparation. This project constitute the dissertation project of Ms.

P. Jayalekshmy who expects to complete her studies here this spring.